Phonon Spectra Prediction in Carbon Nanotubes Using a Manifold-Based Continuum Finite Element Approach

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Massachusetts Institute of Technology Lincoln Laboratory

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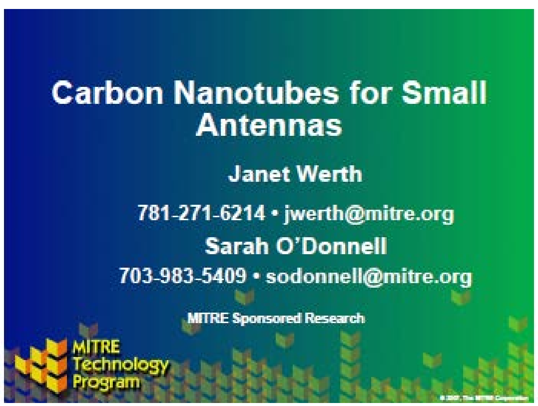
Overview

- CNT Background
- Motivation
- Phonon Behavior
- FEA Development
- Results
- Concluding Remarks



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Phonon spectra prediction in carbon nanotubes using a manifold-based continuum finite element approach

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ABSTRACT

This work develops a tensor-based, reduced-order shell (two-manifold) finite element formulation for predicting phonon spectra in finite-length cylindrical and toroidal carbon nanotubes (CNTs). The formulation does not require an assumed tube thickness. Displacements referencing two covariant basis vectors lying in the tangent space, and one basis vector orthogonal to the tangent space, capture the systems' kinematics. These basis vectors compose a curvilinear coordinate system useful for capturing cylindrical, toroidal, and generically-curved nanotube configurations. The finite element procedure originates from a variational statement (Hamilton's Principle) governing virtual work from internal, external (not considered), and inertial forces. Internal virtual work is related to changes in atomistic potential energy accounted for by an interatomic potential computed at reference area elements. Small virtual changes in the displacements allow a global mass and stiffness matrix to be computed, and these matrices then allow phonon spectra and energies to be predicted via a general eigenvalue problem. Results are generated for example cylindrical and toroidal CNTs documenting accurate prediction of phonon spectra, to include the expected longitudinal, torsional, bending, and breathing-like phonons.

1. Introduction

In order to understand electron scatter and energy transport in a crystalline material, and hence electrical and thermal conductivities, the spectra of phonons (quantized lattice vibration modes) present in the material must first be known. Here, we are interested in the phonon spectra of finite-length carbon nanotubes, and so we employ a semi-classical normal mode description of phonons where each normal mode is a distinct point (frequency and wave vector) on the material's dispersion curve. Follow-on work will reuse the model presented and employ Bloch formalism to compute the complete CNT plane wave dispersion relationships, which characterize fully the phonons in the infinite-length system.

The admissible wave vectors corresponding to each normal mode lead to straight-forward density of state calculations useful in determining the finite-length system's thermal properties [1]. The frequencies also allow ground state energies to be computed, which dictate energy levels below which ballistic electron transport occurs unimpeded by phonon generation. In fact, this consideration motivated the original work – a research team (which includes the two authors) has an interest in using toroidal CNTs [2–4] in a small antenna matching network, but efficiency requirements necessitate operating the CNT in a regime of unimpeded ballistic electron transport.

Analytical approaches exist for predicting phonons present in CNTs, see for example [5]. These methods provide a good first approximation to the actual CNT phonon behavior, including accounting for surface curvature, based on calculations using the planar constitutive CNT material – graphene. Although accurate for idealized lattices, it is unlikely that these techniques could be extended to CNTs with defects, such as the commonly-found Stone Wales defect, due to their reliance on lattice periodicity. The analytical techniques are also expected to provide inadequate results in cases where the CNT exhibits non-idealized geometries such as waviness, sidewall buckling, and large deformation. It is therefore important to develop more-general, computational models for predicting phonon states in such materials. To this end, an approach is presented herein for studying CNT phonon behavior using a continuum finite element formulation. The approach is applied first to cylindrical and toroidal CNTs with perfect crystalline structures. Follow-on work will consider CNTs with defects and non-idealized geometries.

The related study of vibration modes in thin or thick toroidal shells is well-documented and dates to the early 1960's with finite difference solutions presented by McGill and Lenzen ([6,7]). More recent studies have employed the Rayleigh Ritz method [8] to study thin toroidal shells; the differential quadrature method to study thick isotropic toroidal shells [9] and thick orthotropic

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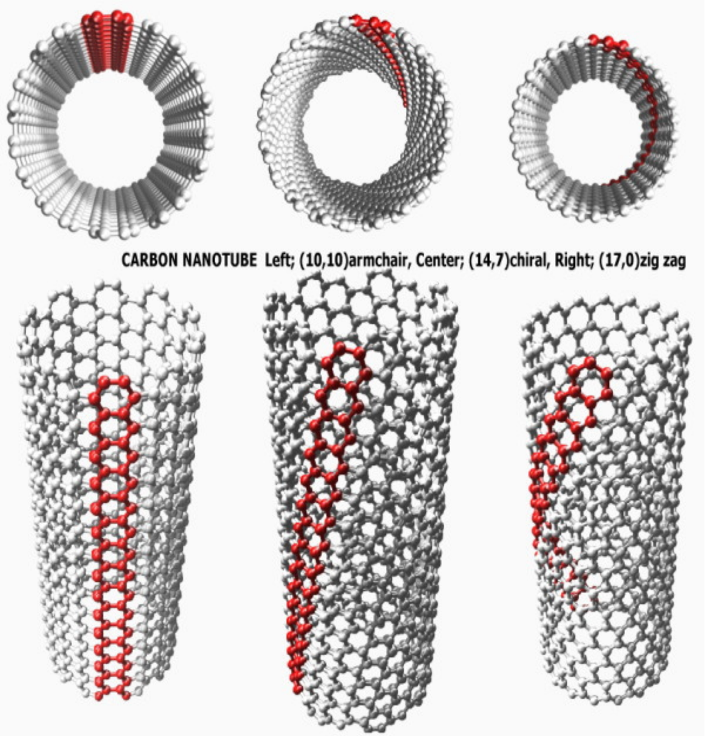
Introduction

- Phonons are quantized lattice vibrations
- Important for understanding thermal, elastic, and electrical properties
- In CNTs, excitation of phonons leads to electron scatter and hence resistance to ballistic electron transport
- This study seeks to quantify phonon spectrum present in CNTs



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CNT Background

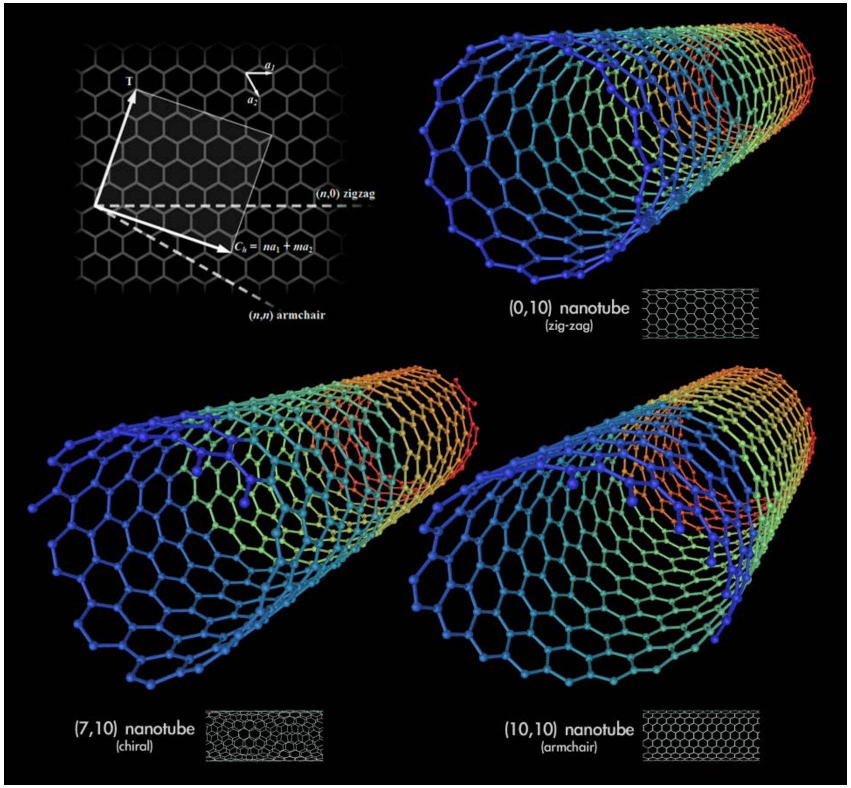
Supposedly discovered in 1991 by S. Iijima, of NEC. Also attributed to 1952 article in Soviet Journal of Physical Chemistry by Radushevich and Lukyanovich.

Length Scale

- **Human hair ϕ ~ 80,000 nm**
- **CNT ϕ ~ 1 nm**

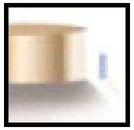
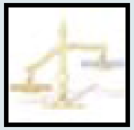
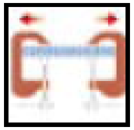
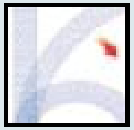
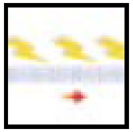
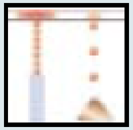
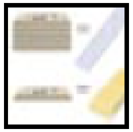
Synthesis

- **Arc discharge**
- **Laser ablation**
- **Chemical Vapor Deposition**



Carbon nanotube - Wikipedia

Motivation – Unique Material Properties!

	PROPERTY	SINGLE-WALLED NANOTUBES	BY COMPARISON
	Size	0.6 to 1.8 nanometer in diameter	Electron beam lithography can create lines 50 nm wide, a few nm thick
	Density	1.33 to 1.40 grams per cubic centimeter	Aluminum has a density of 2.7 g/cm³
	Tensile Strength	45 billion pascals	High-strength steel alloys break at about 2 billion Pa
	Resilience	Can be bent at large angles and restraightened without damage	Metals and carbon fibers fracture at grain boundaries
	Current Carrying Capacity	Estimated at 1 billion amps per square centimeter	Copper wires burn out at about 1 million A/cm²
	Field Emission	Can activate phosphors at 1 to 3 volts if electrodes are spaced 1 micron apart	Molybdenum tips require fields of 50 to 100 V/µm and have very limited lifetimes
	Heat Transmission	Predicted to be as high as 6,000 watts per meter per kelvin at room temperature	Nearly pure diamond transmits 3,320 W/m·K
	Temperature Stability	Stable up to 2,800 degrees Celsius in vacuum, 750 degrees C in air	Metal wires in microchips melt at 600 to 1,000 degrees C
	Cost	$1,500 per gram from BuckyUSA in Houston	Gold was selling for about $10/g in October

P.G. Collis and P. Avouris, "Nanotubes for Electronics,"

Scientific American, Dec. 2000

Discovery Channel – 10 Uses for Carbon Nanotubes

CBCNews

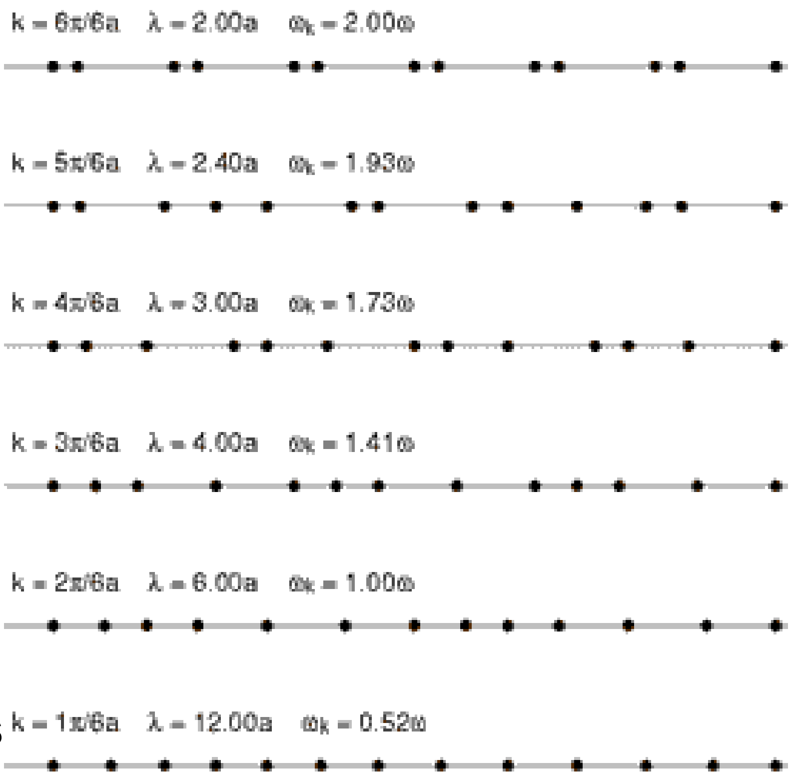
1. Space elevator
2. Faster computer chips
3. Enhanced solar cells
4. Cancer treatment
5. Improved, thinner TVs
6. Better capacitors that replace batteries
7. Flexible displays
8. Bone healing
9. Body armor
10. Faster flywheels

"Theoretically 100 times stronger than steel and six times lighter."

http://dsc.discovery.com/technology/tech-10/carbon-nanotubes-uses.html

Phonon Behavior

Phonon: A phonon is a quantized mode of vibration occurring in a rigid crystal lattice, such as the atomic lattice of a solid.



Classical Approach: modal analysis

$$\mathrm{M}_{\mathrm{IJ}}^{\sigma\rho}\ddot{\hat{U}}_J^{\rho} + \mathrm{K}_{\mathrm{IJ}}^{\sigma\rho}\hat{U}_J^{\rho} = 0$$

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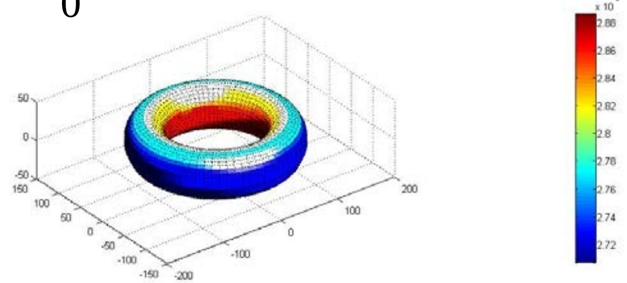
Modeling Approach

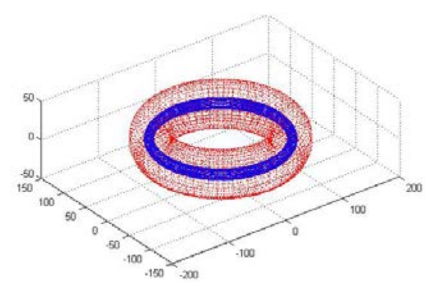
Derive shell-like equations of motion referencing basis vectors in the undeformed tangent space

Equate strain energy to interatomic potential energy

Discretize EOMs using finite elements $T_X \Omega_0$

Solve for normal modes of vibration





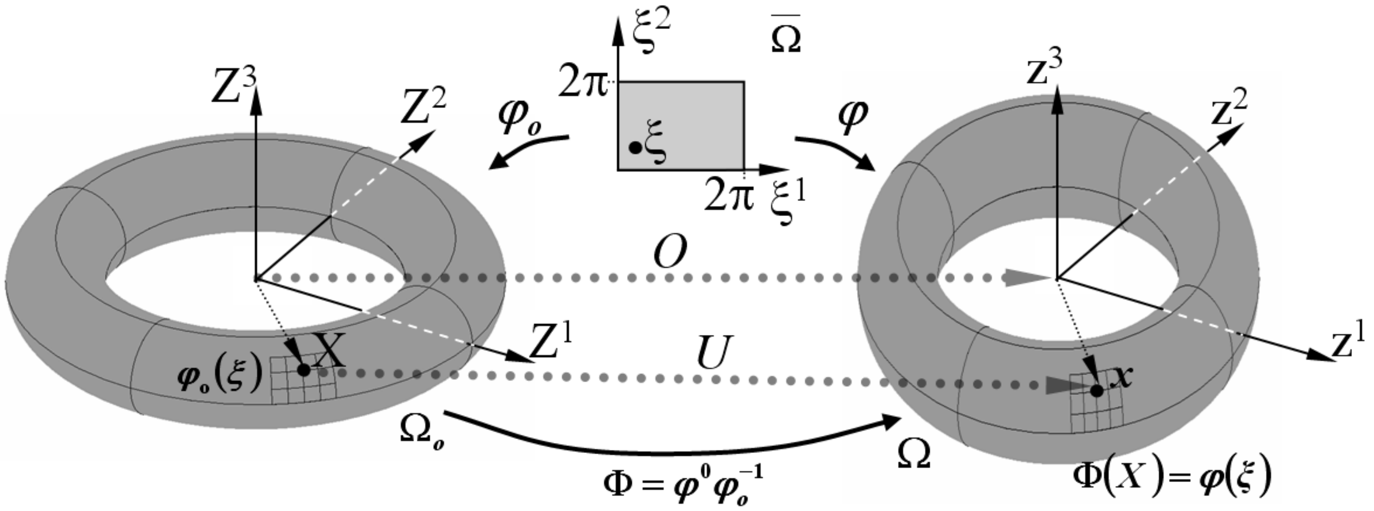
Model - Kinematics

Mappings from parametric body

Vectors referencing tangent spaces and their basis vectors

$$\boldsymbol{G}_\alpha \equiv \frac{\partial \boldsymbol{X}}{\partial \xi^\alpha} = \frac{\partial Z^a}{\partial \xi^\alpha}\boldsymbol{I}_a$$

$$\boldsymbol{g}_\alpha \equiv \frac{\partial \boldsymbol{x}}{\partial \xi^\alpha} = \frac{\partial z^a}{\partial \xi^\alpha}\boldsymbol{i}_a$$



$$\Phi = \varphi \circ \varphi_0^{-1} : X \in \Omega_0 \mapsto x = \Phi(X) = \varphi(\varphi_0^{-1}(X)) \in \Omega$$

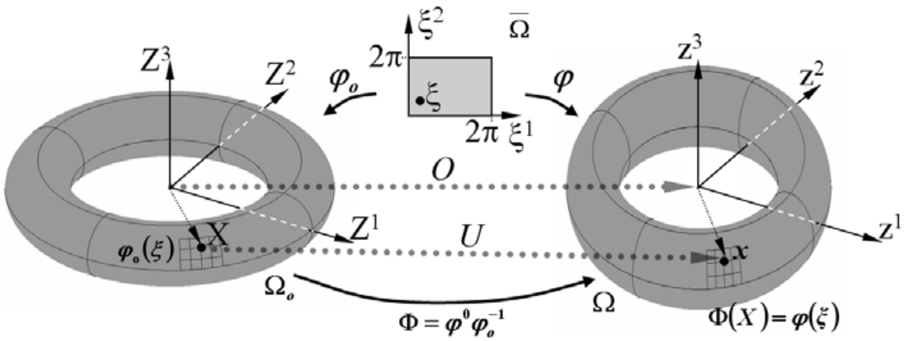
$$\Phi : \boldsymbol{X} \mapsto \boldsymbol{x} = \boldsymbol{\Phi}(X) = \boldsymbol{X} + \boldsymbol{U}(X) - \boldsymbol{O}$$

$$\boldsymbol{g}_\alpha = \boldsymbol{G}_\alpha + \frac{\partial \boldsymbol{U}}{\partial \xi^\alpha} = \boldsymbol{G}_\alpha + \frac{\partial U^\beta}{\partial \xi^\alpha}\boldsymbol{G}_\beta + U^\beta \frac{\partial \boldsymbol{G}_\beta}{\partial \xi^\alpha}$$

Model - Kinematics

Deformation gradient maps line segments in tangent spaces

$$\mathbf{F} \equiv \frac{\partial \boldsymbol{x}}{\partial \boldsymbol{X}} \in \mathrm{R}^{3\times 3} : T\Omega_0 \to T\Omega \qquad \boldsymbol{dx} = \mathbf{F} \cdot \boldsymbol{dX}$$



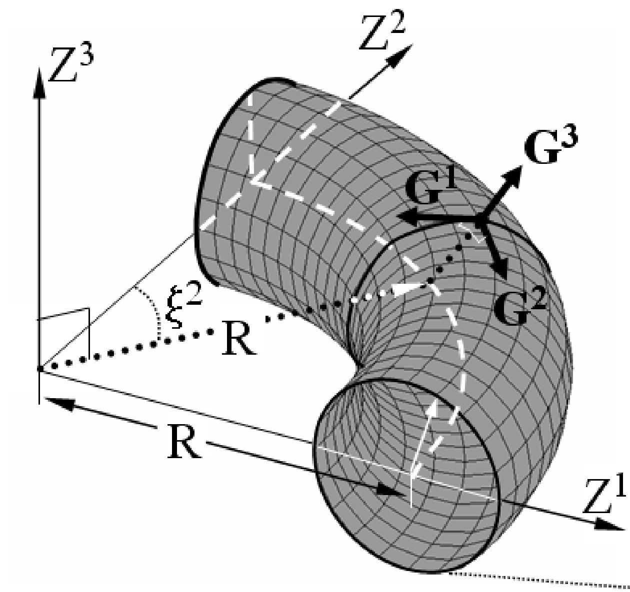
Gradient

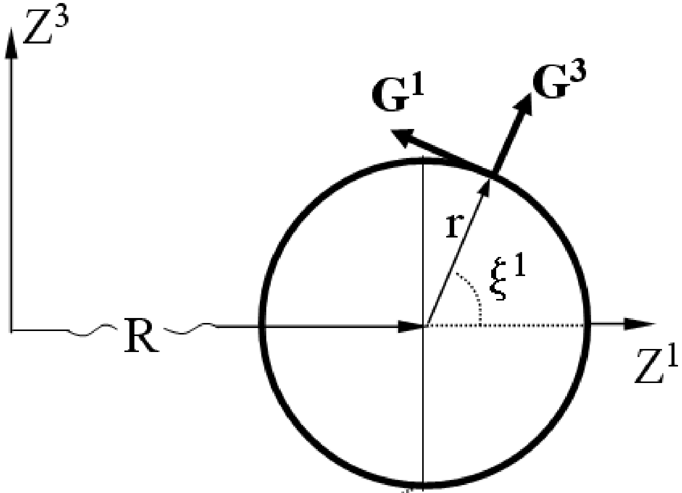
$$\mathbf{F} \equiv \frac{\partial \boldsymbol{x}}{\partial \boldsymbol{X}} = \frac{\partial \boldsymbol{x}}{\partial \xi^{\alpha}} \frac{\partial \xi^{\alpha}}{\partial \boldsymbol{X}} = \boldsymbol{g}_{\alpha} \otimes \boldsymbol{G}^{\alpha} = \delta^{\alpha}_{\beta} \boldsymbol{g}_{\alpha} \otimes \boldsymbol{G}^{\beta}$$

$$\nabla_X \equiv \frac{\partial}{\partial \boldsymbol{X}} = \frac{\partial}{\partial \xi^{\alpha}} \frac{\partial \xi^{\alpha}}{\partial \boldsymbol{X}} = \boldsymbol{G}^{\alpha} \frac{\partial}{\partial \xi^{\alpha}} = G^{\alpha\beta} \boldsymbol{G}_{\beta} \frac{\partial}{\partial \xi^{\alpha}}$$

Parameterization

Toroidal Parameterization





$$Z^1 = (R + \xi^3 \cos\xi^1)\sin\xi^2$$

$$Z^2 = (R + \xi^3 \cos\xi^1)\cos\xi^2$$

$$Z^3 = \xi^3 \sin\xi^1$$

$$r = \xi^3$$ **Constant!**

Yields G_α and Christoffel Symbols $\Gamma^\eta_{\rho\beta}$

$$\mathbf{F} \equiv \frac{\partial \boldsymbol{x}}{\partial \boldsymbol{X}} = \left[\delta^\eta_\beta + \frac{\partial U^\eta}{\partial \xi^\beta} + U^\rho \Gamma^\eta_{\rho\beta}\right] \boldsymbol{G}_\eta \otimes \boldsymbol{G}^\beta$$

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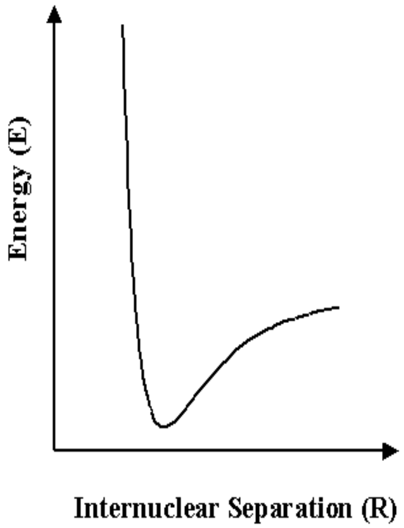
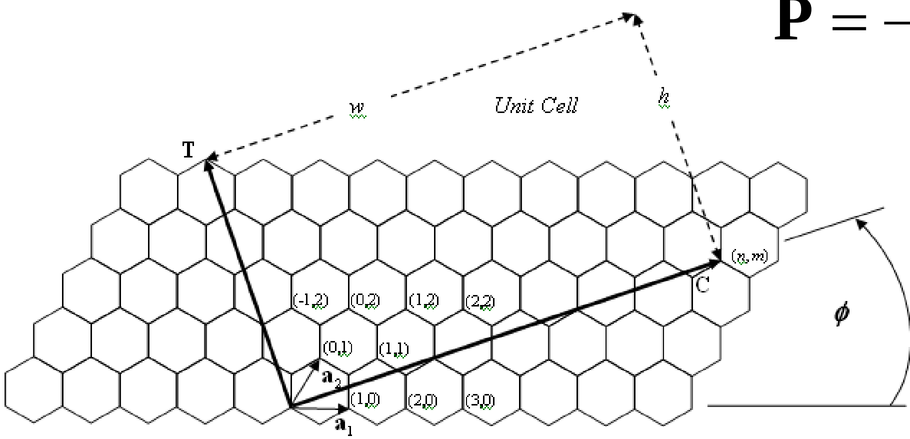
Energy Connection

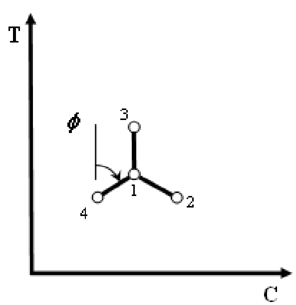
$$W_0 \equiv \frac{E^{rae}}{A_{Hex}} = \frac{E^{rae}_{stretch} + 2E^{rae}_{angle}}{A_{Hex}}$$

$$\mathbf{P} = \frac{\partial W_0}{\partial \mathbf{F}}$$





Graphene Sheet



Reference Area Element

- **Bond vectors**

$$\boldsymbol{r}_{ij} = \mathbf{F} \cdot \boldsymbol{R}_{ij}$$

- **Modified Morse Potential**

$$E_{stretch} = D_e \left(\left[1 - e^{-\beta(r - r_0)} \right]^2 - 1 \right)$$

$$E_{angle} = \frac{1}{2} k_\theta (\theta - \theta_0)^2 \left[1 + k_{sextic} (\theta - \theta_0)^4 \right]$$

Equations of Motion

Hamilton's Principle

$$\mathbf{P} = \frac{\partial W_0}{\partial \mathbf{F}}$$

$$\mathrm{P}_{\alpha}^{\beta} = \mathrm{P}_{\alpha}^{\beta}(\boldsymbol{F}) = \frac{\partial W_0}{\partial \mathrm{F}_{\beta}^{\alpha}} = \frac{\partial W_0}{\partial r_{1k}} \frac{\partial r_{1k}}{\partial \mathrm{F}_{\beta}^{\alpha}} + \frac{\partial W_0}{\partial \theta_{1kl}} \frac{\partial \theta_{1kl}}{\partial \mathrm{F}_{\beta}^{\alpha}}$$
$$= \frac{1}{A^{rae}} \left(\frac{\partial E^{rae}}{\partial r_{1k}} \frac{\partial r_{1k}}{\partial \mathrm{F}_{\beta}^{\alpha}} + 2 \frac{\partial E^{rae}}{\partial \theta_{1kl}} \frac{\partial \theta_{1kl}}{\partial \mathrm{F}_{\beta}^{\alpha}} \right),$$

$$\delta E^{tot} = \delta \int_{\Omega_0} W_0(\mathbf{F})\, d\Omega_0 + \delta E^{ext} = \int_{\Omega_0} \frac{\partial W_0}{\partial \mathbf{F}} : \delta \mathbf{F}\, d\Omega_0 + \delta E^{ext} = 0$$

where

$$\delta E^{ext} = -\int_{\Omega_0} \delta \boldsymbol{U} \cdot \boldsymbol{t}_0^P\, d\Omega_0 + \int_{\Omega_0} \rho_0 \ddot{\boldsymbol{U}} \cdot \delta \boldsymbol{U}\, d\Omega_0$$

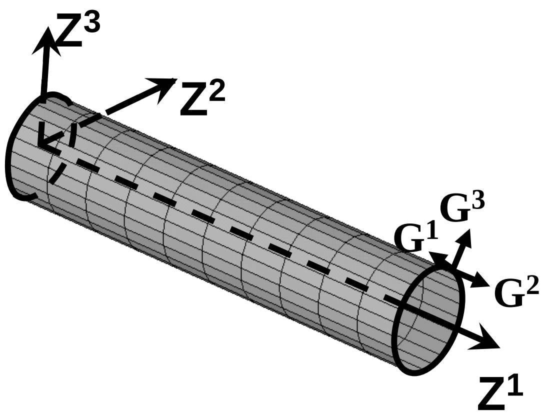
0

Interpolate *U*

$$\boldsymbol{U} = N^I(\xi^1, \xi^2)\, \hat{U}_I^i \bar{t}_i^{\beta} \boldsymbol{G}_{\beta}$$

Taylor expand P

$$\mathrm{P}_{\alpha}^{\beta} = \mathrm{P}_{\alpha}^{\beta 0} + \frac{\partial^2 W_0}{\partial \mathrm{F}_{\beta}^{\alpha} \partial \mathrm{F}_{\sigma}^{\mu}} \frac{\partial \mathrm{F}_{\sigma}^{\mu}}{\partial \hat{U}_J^{\rho}} \Bigg|_{\boldsymbol{U}=0} \hat{U}_J^{\rho} + O(\hat{U}_J^{\rho\,2})$$



Expressions found in closed form

Equations of Motion

Stiffness matrix from interatomic energy

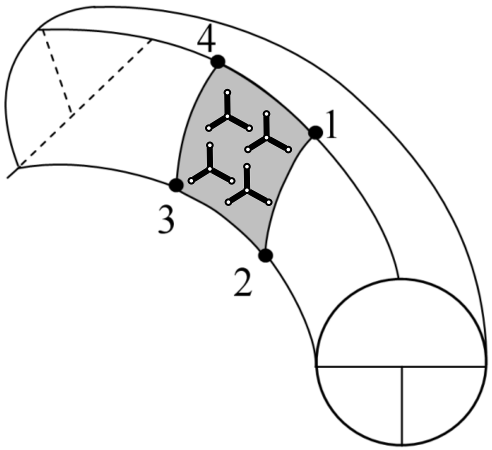
$$\mathrm{K}_{\mathrm{IJ}}^{\sigma\rho} = \int_{\Omega_0} \left(\frac{\partial^2 W_0}{\partial \mathrm{F}_\beta^\alpha \partial \mathrm{F}_\sigma^\mu} \frac{\partial \mathrm{F}_\sigma^\mu}{\partial \hat{U}_J^\rho} \frac{\partial \mathrm{F}_\beta^\alpha}{\partial \hat{U}_I^\sigma} \right) d\Omega_0$$

Mass matrix from inertial work

$$\mathrm{M}_{\mathrm{IJ}}^{\sigma\rho} = \int_{\Omega_0} \rho_0 N^I N^J \bar{t}_\sigma^i \bar{t}_\rho^j G_{ij} d\Omega_0$$

Completed form of EOMs

$$\mathrm{M}_{\mathrm{IJ}}^{\sigma\rho} \ddot{\hat{U}}_J^\rho + \mathrm{K}_{\mathrm{IJ}}^{\sigma\rho} \hat{U}_J^\rho = 0$$



4-noded shell element used in all results presented

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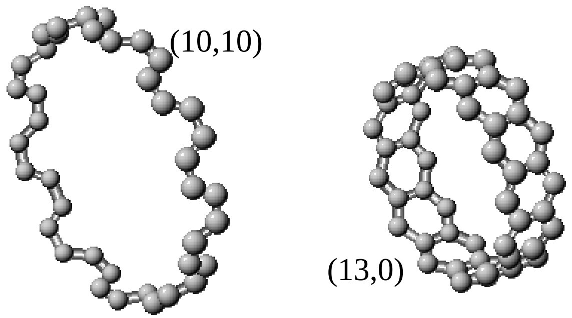
Results - RBM Validation



Radii: 6.765 Angs. 5.08 Angs.

Experimentally Determined Values[1]

(10,10) : 4.86 THz

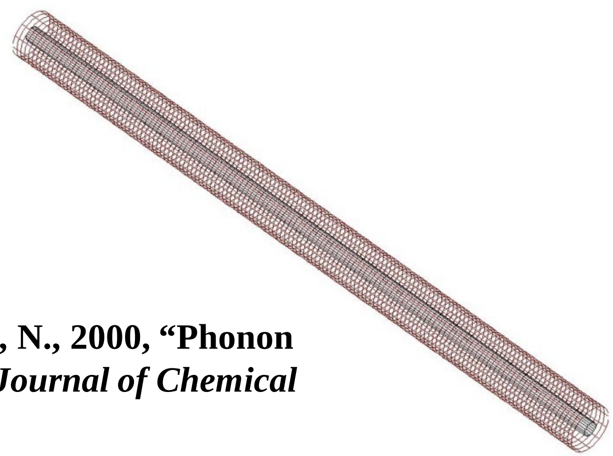
(13,0) : 6.33 THz

Quantum Calculations[1]

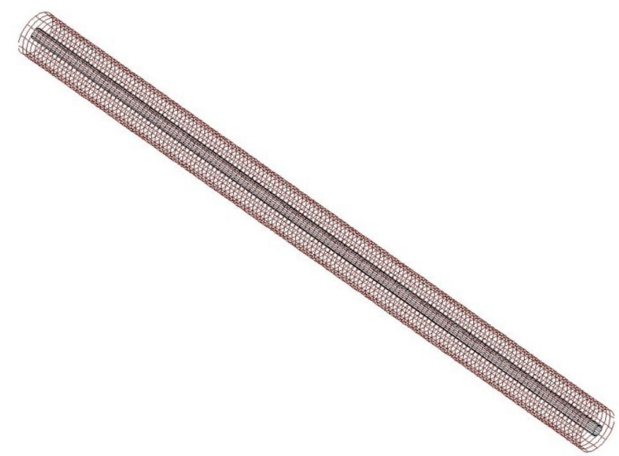
(10,10) : 4.7 - 5.85 THz

(13,0) : 7.41 THz

5.2682 THz



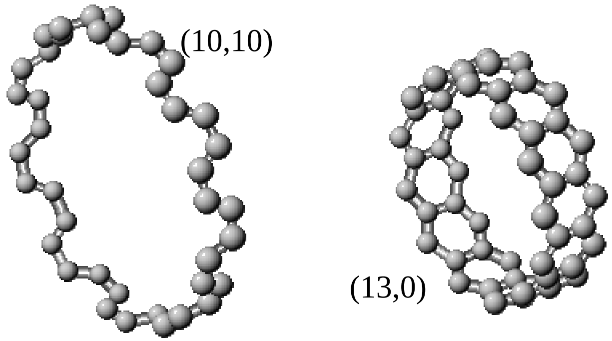
7.0002 THz



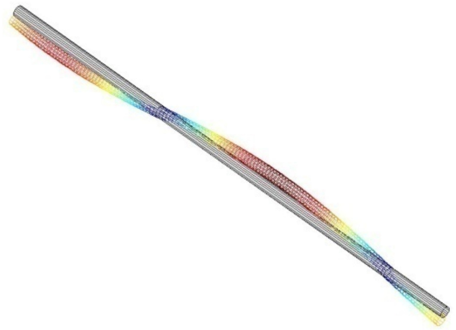
[1] Sokhan, VP, Nicholson, D., Quirke, N., 2000, “Phonon spectra in model carbon nanotubes,” *Journal of Chemical Physics*, 113(5), pp 2007-2015.

(10,10) Radial Breathing Mode **(13,0) Radial Breathing Mode**

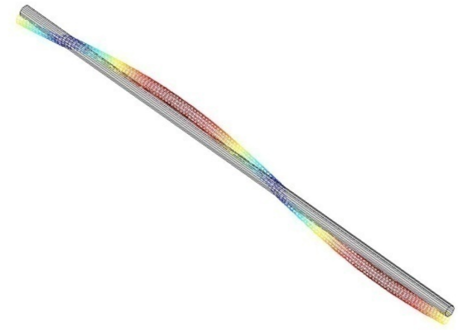
Results – (10,10) Bending



40.9262 GHz



41.2959 GHz



Radii: 6.765 Angs. 5.08 Angs.

45.438 GHz

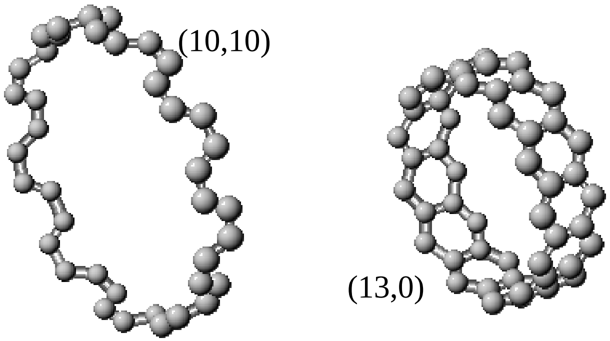
45.7258 GHz

95.3129 GHz

Other methods[2] yield 41.2 GHz as fundamental bending mode

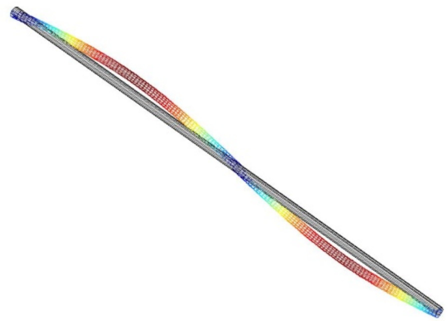
[2]Srivastava, D., Wei, C., Kyeongjae, C., 2003, "Nanomechanics of carbon nanotubes and composites, " Applied Mechanics Reviews 56 (2), pp. 215-230.

Results – (13,0) Bending

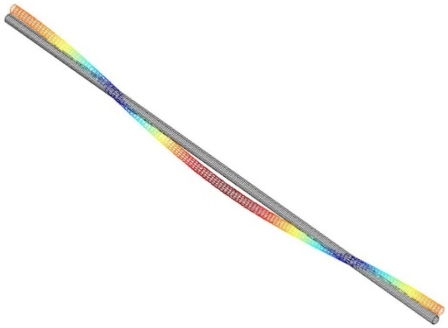


Radii: 6.765 Angs. 5.08 Angs.

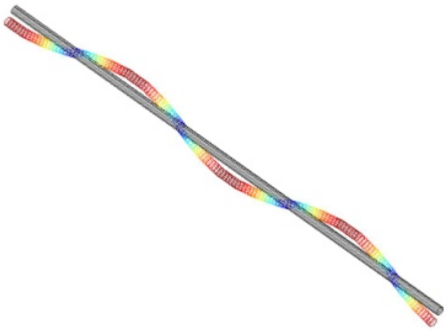
14.9898 GHz



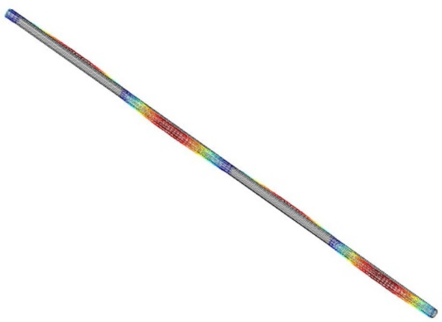
17.3485 GHz



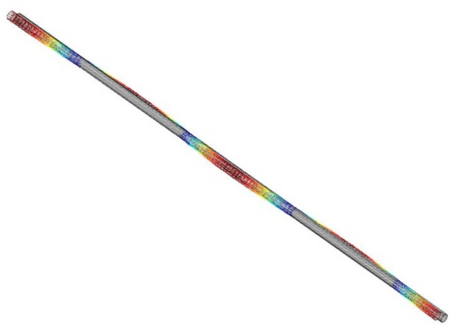
81.872 GHz



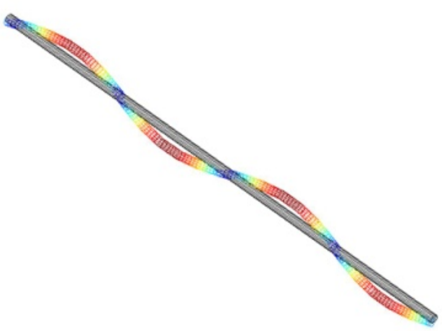
78.3678 GHz



79.2083 GHz

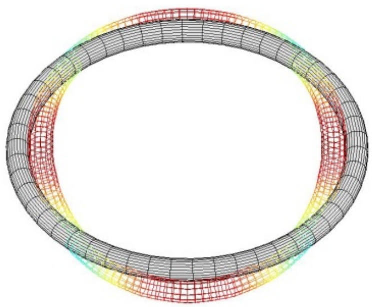


82.7701 GHz

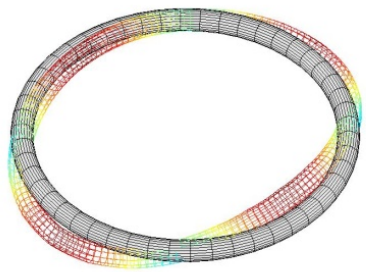


Results – (10,10) Toroidal Bending

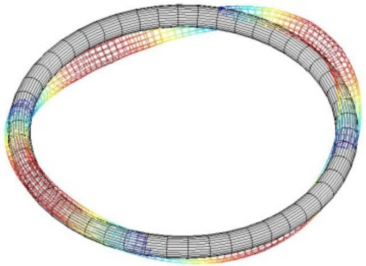
60.1693 GHz



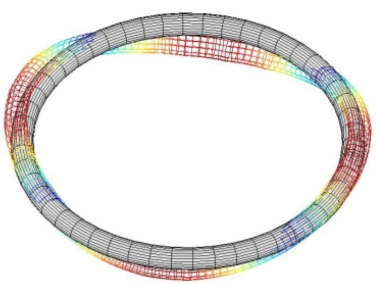
62.3895 GHz



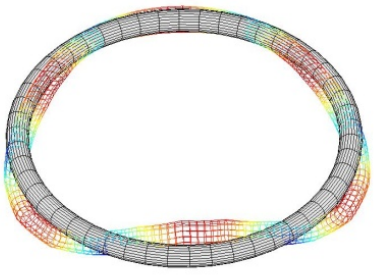
65.5137 GHz



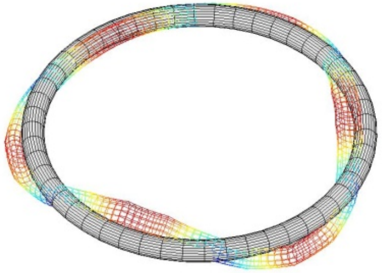
65.6843 GHz



158.0734 GHz



158.1115 GHz



Concluding Remarks

- CNT have many outstanding properties
- Finite element formulations have been developed for predicting phonon spectra
- The method compares favorably with experimental results and computationally intensive algorithms
- Models generalize well to imperfect tubes and non-ideal geometries (wavy, etc.)
- Elements are re-usable

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ACKNOWLEDGEMENTS

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Top 5 Uses

1. [Space elevator]
Sending a payload into space by rocket is expensive ($10,000 per pound) and dangerous. Some are proposing a very tall elevator that would stretch from the ground to beyond Earth's atmosphere. Making this a reality requires a long, strong cable tethered to a counterweight in geosynchronous orbit maintained at a fixed position about 22,000 miles above the earth. CNTs are the only known material up to the task. Among other things, a successful space elevator could be a means for safe disposal of nuclear waste and give life to a space tourism industry.

2. Faster computer chips
The processing speed of a computer chip depends on the number of transistors it has. Today, typical desktop processors using silicon transistors have less than half a billion. Computer chips using CNTs could blow that number away. Their small size - just one nanometer wide - means many billions of CNT transistors could be packed onto a single processing chip making for smaller, faster computers and electronics.

3. Better solar cells
Semiconducting materials altered with certain impurities are used in solar cells. When struck by light, these materials release electrons, creating usable electricity. Most of today's solar cells use silicon semiconductors, but that could change. Because they're so tiny, billions of CNTs could be tightly packed onto solar cells and release far more electricity per square inch than silicon. In addition, carbon nanotubes absorb light so well that a professor at Rice University used them to create the darkest ever man-made material.

4. Cancer treatment
CNTs are so small they might one day be used to target and destroy individual cancer cells. By treating CNTs with certain proteins, scientists are developing a method to bind them specifically to cancerous cells. Once attached, the CNTs, which are excellent conductors of heat, could be exposed to infrared light shone through the patient's skin. The light would heat the CNTs to a temperature high enough to destroy the cancer cells while leaving surrounding tissue undamaged. While more research must be done, this method could offer a way to treat certain cancers without harming healthy tissue, a current drawback of treatments like chemotherapy.

5. Better, thinner TVs
Traditional tube TVs essentially work by firing electrons at substances called phosphors to make them glow, creating the colored light of a television picture. This process requires an electron gun in a relatively big picture tube. But new displays, called field emission displays miniaturize the process by using tiny electron emitters positioned behind individual (microscopic) phosphorus dots. An array of CNTs, which are excellent electron emitters, could be used in field emission displays to excite the phosphorus dots, creating bright, high resolution displays that are only millimeters thick and consume less power than plasma and liquid crystal displays.

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Top 6-10

6. Better capacitors that replace batteries
Instead of storing electricity chemically like a battery, capacitors hold it physically by building a charge on a material called a dielectric. The dielectric's surface area determines how much charge it can hold. CNTs have extraordinarily high surface areas, and using them as the dielectric could increase the storage ability of capacitors to be on par with modern batteries. But if we already have batteries what's the use? Batteries take hours to charge and lose their capacity with time. Capacitors don't have these problems. CNT capacitors might one day be used in instantly rechargeable laptops and electric cars.

7. Flexible displays
The dream of fold-up TVs and computer screens that can fit inside people's pockets has, up until now, been stifled by rigid silicon semiconductors. While some organic semiconductors have used in bendable plastic displays, their performance has been fairly poor. But CNTs, in addition to being very flexible, compare favorably to silicon in terms of performance. Researchers at Purdue and the University of Illinois-Urbana-Champaign are developing carbon nanotube flexible displays which one day could be used for things like electronic newspapers and roll-up handheld devices.

8. Bone healing
Researchers at the University of California-Riverside have discovered that CNTs can act as scaffolds around which bone cells will grow by attracting hydroxyapatites, calcium crystals in the body that are critical for bone formation. The technology might one day help individuals with bone diseases or particularly catastrophic injuries regrow bones.

9. Body armor
Researchers at Cambridge University have figured out how to spin many tiny carbon nanotubes together to create fibers that have the strength of Kevlar, a composite material used in bullet-proof vests. With new techniques rapidly emerging to make longer CNTs, spun fibers using the longer CNTs will soon surpass Kevlar in strength, and weigh less. As CNT prices drop, spun CNT fibers could be the material of choice for better, lighter body armor.

10. Faster flywheels
A flywheel is like a battery in that it stores energy. But unlike a battery, this energy is mechanical and stored via a wheel rotating at high speed (the faster the spin, the more energy it stores). Flywheels offer certain advantages over batteries. But a flywheel, if spun too fast, can shatter because of the strength limits of its material. Because of their strength, CNTs could be used to make faster flywheels that store more energy without shattering. While Flywheels have seen only limited use in amusement rides, race cars and backup power supplies, using CNTs could allow flywheels to become more prevalent in areas like public transportation and hybrid cars.

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